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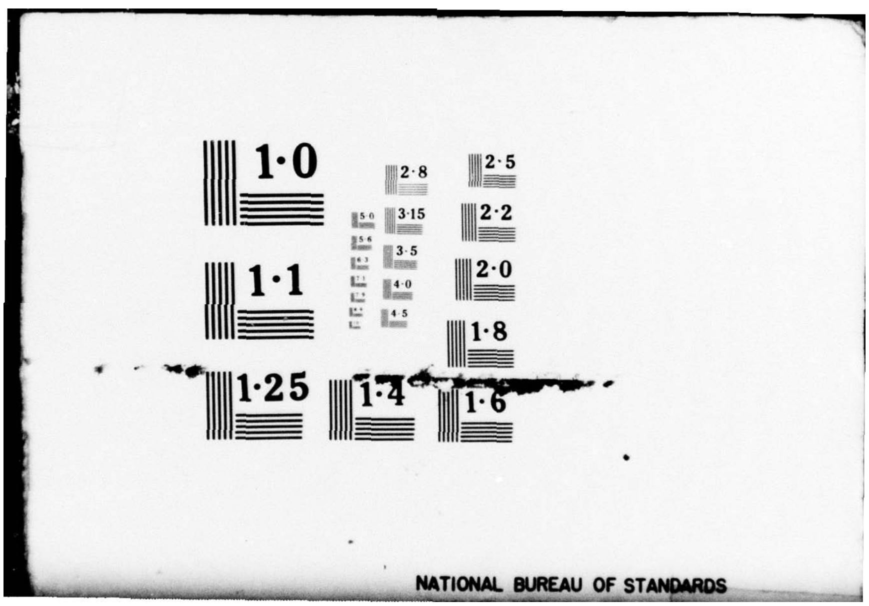
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RE-ENTRANT SUPERCONDUCTIVITY IN A MAGNETICALLY ORDERED SUPERCONDUCTOR: $La_{1-x}Gd_xRu_2$

T.E. Jones,[redacted] J.F. Kwak,† E.P. Chock [redacted] P.M. Chaikin

Department of Physics,* University of California, Los Angeles, California 90024, USA

We have measured the critical temperature (T_c) and the upper critical magnetic field (H_{c2}) of $La_{1-x}Gd_xRu_2$. At low concentrations of the magnetic impurity (Gd), the suppression of T_c follows the expected Abrikosov-Gorkov (A-G) pair breaking curve. However, for larger concentrations, strong deviations below A-G are observed. Samples in this region ($4. \lesssim x \lesssim 5.$ at. %) exhibit two T_c's. $La_{1-x}Gd_xRu_2$ is known to order magnetically, probably as a spin glass, and the magnetic ordering temperature (T_M) has been measured in the normal state. This T_M curve intersects the T_c curve in the concentration range where the T_c curve is re-entrant and we therefore attribute the re-entrant T_c behavior to the magnetic ordering of the Gd^{3+} ions.

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The destructive effect of magnetic impurities and of applied magnetic fields, on superconductivity, has been of considerable interest[1-3]. Except for the compensation effect[4-5], increased doping with magnetic impurities and large internal exchange fields always have harmful effect on superconductivity. The question of whether superconductivity and any type of magnetism can co-exist, has been debated for many years.[6] No known ferromagnetic material becomes superconducting upon cooling to any temperature. However, some superconductors, when cooled below their transition temperature (T_c), do undergo a magnetic transition. In previously studied systems a variety of behaviors have been observed in T_c vs. concentration and H_{c2} curves.[7]

$(La_{1-x}Gd_x)Ru_2$ was chosen for this study for several reasons. The phase diagram as investigated by Hillenbrand and Wilhelm[8] indicates a magnetic ordering temperature (T_M) vs. concentration curve which intersects the T_c curve between 4 and 5 atomic percent Gd at which point the T_c is still at a reasonably high temperature (approximately 2K). More recent work[5,9,10] has shown that the internal exchange field of the Gd ions is large ($J \approx -.015ev$) and that the system is exchange enhanced. In this paper we have undertaken a more detailed study of the T_c versus concentration (x) behavior in this system in the region of concentration, 4.-5. at.% Gd, where the two phase boundary curves meet.

Our first hint that something really spectacular might be occurring in this system, came from our critical field data. In reference 5 for $(La_{.993}Lu_{.007})_{1-x}Gd_xRu_2$, we found that the critical field curve for the highest concentration re-entrant H_{c2} samples almost return to the temperature axis at the lowest temperatures. If this occurred, it would mean that this material was re-entrant in the T_c-concentration plane. Such behavior has been observed previously only in Kondo systems.[11] This had never been observed in a magnetically ordered superconductor before.

In figure 1, some interesting re-entrant T_c traces are illustrated. Note, in zero field, there is a range of approximately 1.3°K where the sample is superconducting. Heating above this temperature range, or cooling below it, would return the material to the normal state. The latter event is extraordinary.

Both transitions are much sharper than has been observed in the Kondo systems, the transition width being ≈20 mK. This was one of the first observations of the disappearance of superconductivity at lower temperatures in a non-Kondo system.[12] We attribute this striking behavior to the magnetic ordering of the Gd in this system. This will be discussed more below. Note further in figure 1, as a larger magnetic field is applied the upper transition, T_{c1}, is lowered and the lower transition, T_{c2}, is raised. That is, the region of superconductivity is reduced from both sides. When a field of 250 gauss is applied, the material does not go completely superconducting. When a 500 gauss field is applied, there is no sign of any superconductivity at all.

The same re-entrant T_c behavior has been observed in samples of different concentrations, as illustrated in figure 2. Note, as T_{c1} is reduced, T_{c2} increases and the temperature range when the sample is superconducting is narrowed for higher concentrations. This behavior is similar to that illustrated in figure 1, when a magnetic field is applied to the system.

Our T_c versus concentration data is summarized in figure 3. The T_c behavior in our Lu-doped[13] system should be compared with reference 8. The present data is clearer; the transition temperatures are very reproducible. For high concentrations at the re-entrant end of the curve, both Lu-doped and pure $LaRu_2$ samples exhibit re-entrant superconductivity and they seem to fit the same curve, as far as we can determine.

The data shown in figures 1, 2 and 3 were also measured inductively and showed flux expulsion and the presence of two transitions in very good agreement with the resistance measurements. The inductive measurements were complicated however by the temperature and field dependent susceptibility of the Gd spin glass system.[14] Specific heat measurements were attempted on several samples but were dominated by the spin glass effects.[15] The superconducting specific heat jump decreased rapidly with increasing Gd concentration. With our present sensitivity, the 4% Gd sample had a barely perceptible change at T_c. For $x \lesssim 4\%$ the specific heat measurements yielded T_c and H_{c2}, in excellent agreement with the resistance measurements.

The hysteresis indicated in figures 1 and 2 represents a 20 mK temperature width in zero field. This increases to about 40 mK as the magnetic field increases. This hysteresis was measured very carefully with two different probes, a dilution refrigerator and a He^3 cryostat (where the sample was in direct contact with a He^3 bath) and by two different people. The hysteresis is real and not the result of any thermal gradients in the sample chamber We thought, at first, that this hysteresis might be indicative of a first order transition. However, specific heat measurements in our laboratory have not detected this transition to the sensitivity of the apparatus.

The concentrations used in figure 3 have been determined as well as possible, without the benefit of a precise chemical or physical analysis. The concentrations used were usually the nominal concentrations, as advertised by the manufacturer, E. P. Chock. Samples made to be 4.9 or 5.0 at. % Gd or higher, never went superconducting. This puts an upper limit of approximately 5 at. % on the critical concentration. Samples made to be 4.4, 4.6 and 4.8 at. %, those initially measured, were used to define the position of the T_{c1} versus x curve. Subsequently, data which showed a small amount of scatter was fitted to this T_{c1} curve. In all cases, the value of T_{c2} shown was graphed at the concentration determined by the T_{c1} of the sample. Thus, the absolute accuracy of the position of the high concentration end shown in figure 3 may be in error by several tenths of a percent of Gd concentration. However, the nesting of the points is unmistakable. It can be said with certainty that samples with a Gd concentration such that their T_{c1} is reduced always have their T_{c2} increased, as shown in figure 3.

The precise nature of the magnetic order in this system has not been determined. It seems that a spin glass type of magnetic order is most likely, due to the random positions of the magnetic ions in such an alloy system. A spin glass has a cusp or peak in the susceptibility at T_M, which washes out and may be lowered in temperature as a magnetic field is applied. Our system exhibits this behavior as indicated by our inductance measurements which are similar to those found previously in $LaRu_2$, $ThRu_2$, and $CeRu_2$ doped with Gd.[8,10] The suggestion that these systems are spin glass superconductors has previously been made by Davidov et al.[16] Furthermore, evidence for spin glass behavior comes from specific heat measurements made in our laboratory which indicate the characteristic temperature dependence found by Peter et al.[15]

A summary of our current knowledge of the T_c and T_M behavior of $(La_{1-x}Gd_x)Ru_2$ is illustrated in figure 4. We have determined the T_c versus x curve much more precisely than it had been previously, especially in the region between 4.0 and 5.0 at.% Gd, where the re-entrant nature of the curve has been elucidated. The T_c versus x curve turns around in the region where the magnetic ordering curve meets the superconducting curve. We therefore attribute the re-entrant superconductivity to magnetic ordering of the Gd^{3+} ions in the system.

More recently there have been reports on re-entrant transition temperatures resulting from magnetic ordering in the superconducting compounds $ErRh_4B_4$[17] and $Ho_{1.2}Mo_6S_8$[18]. In these systems the second superconducting transition, marking the return to the normal state as the temperature is lowered, appears to be coincident with the magnetic ordering temperature. This may be the result of a rather weak coupling between the magnetic and superconducting subsystems. $LaRu_2$ doped with Gd has a phase diagram (figure 4) in which the magnetic ordering curve extrapolates into the superconducting region. We may therefore have a region of coexistence of superconductivity and magnetic order, but this hypothesis requires more experimental work. From the low concentration part of the phase diagram we believe that $La_{1-x}Gd_xRu_2$ should be considered an A-G system with multiple pair-breaking from spin flip scattering and internal exchange fields both of which are temperature and concentration dependent.

In the previously studied Kondo superconductors, the increased pair-breaking which results in two T_c's comes from an anomalous increase in the exchange scattering rate, $1/\tau_s$, at the temperature is lowered. Although this is an interesting effect, it is quite different from what we are seeing. In $La_{1-x}Gd_xRu_2$, and presumably in the other two magnetic superconductors mentioned above, there is a well defined long lived local moment which "freezes" in place below the magnetic ordering temperature. This precludes an increase in the spin-flipping exchange scattering, because the spins frozen in place are not free to flip. Therefore, a magnetically ordered system, such as ours, cannot exhibit Kondo effects. In fact, Gd does not show a Kondo effect in any host, as far as we know.

We would therefore suggest that the reentrant T_c behavior reported in this paper is a result of the presence of a substantial internal exchange field which develops below T_M of the Gd spins. We have seen large internal field effects at lower concentrations.[5,9] Note, that for 5 at.% Gd doping the fully aligned spins would produce an effective field of 225,000 gauss. A small fraction of spin alignment would thus produce a sufficient field to quench the superconductivity of $LaRu_2$ which has a Pauli critical field of only about 50,000 gauss in the absence of any other pair-breaking.[5] Although a spin glass should strictly speaking have zero net magnetization there may be strong deviations from zero on the volume scale of a coherence length cubed. This may result either from statistical fluctuations or from the nature of the short range order which is not well understood. More work is needed on the forms of the magnetic ordering in this compound.

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REFERENCES

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† Present address: Naval Ocean Systems Center, San Diego, California 92152.

‡ Present address: IBM Research Labs, San Jose, California 95193.

1. MATTHIAS, B.T., SUHL, H. and CORENZWIT, E., Phys. Rev. Lett. 1, 92 (1958).
2. ABRIKOSOV, A.A. and GOR'KOV, L.P., Soviet Phys. JETP 15, 752 (1962).
3. FULDE, P and MAKI, K., Phys. Rev. 141, 275 (1965).
4. FISCHER, O.H., Heletica Physica Acta 45, 331 (1972).
5. JONES, T.E., CHOCK, E.P. and CHAIKIN, P.M., Solid State Comm. 20, 635 (1976).
6. GOR'KOV, L.P. and RUSINOV, A.I., Soviet Phys. JETP 19, 922 (1964).
7. PARKS, R.D., Superconductivity, Vol. 2, WALLACE, P.R. (ed.) Gordon and Breach, N.Y.(1969).
8. HILLENBRAND, B. and WILHELM, M., Phys. Lett. 40A, 387 (1972).
9. RETTORI, C., DAVIDOV, D., CHAIKIN, P., and ORBACH., R., Phys. Rev. Lett. 30, 437 (1973).
10. ENGEL, U., Thesis, University of Berlin, 1974 (unpublished).
11. MAPLE, M.B., Magnetism V (ed. by Rado and Suhl), Academic Press, New York (1973).
12. JONES, T.E., CHOCK, E.P. and CHAIKIN, P.M., (Bull. Am. Phys. Soc. 22, 339 (1977).
13. In our previous studies of $LaRu_2$ (ref. 5) we have found that doping with a small concentration of Lu yields much more reproducible results for T_c and H_{c2}. We believe this is due to a suppression of the martensitic transition at ~30K. For high Gd concentration we believe the structure is stabilized by the Gd itself.
14. CANNELLA, V. and MYDOSH, J.A., Phys. Rev. B6, 4220 (1972).
15. PETER, M., DONZE, P., FISCHER, O., JUNOD, A., ORTELLI, J., TREYVAUD, A., WALKER, E., WILHELM, M. and HILLENBRAND, B., Helv. Phys. Acta 44, 345 (1971).
16. DAVIDOV, D., BABERSCHKE, K., MYDOSH, J.A. and NIEWENHUYS, G.J., J. Phys. F7, L47 (1977).
17. FERTIG, W.A., JOHNSTON, D.C., DELONG, L.E., McCALLAM, R.W., MAPLE, M.B., and MATTHIAS, B.T., Phys. Rev. Lett. 38, 987 (1977).
18. SHIKAWA, M.I. and FISCHER, O., to be published.

FIGURE CAPTIONS

Fig. 1. The normalized 4 probe resistance is plotted versus temperature in several magnetic fields. The arrows on the lower transision represent the direction of temperature change and the observed hysteresis.

Fig. 2. Normalized resistance vs. temperature for $(La_{1-x}Gd_x)Ru_2$, for several different concentrations of Gd.

Fig. 3. Transition temperature vs. concentration of Gd. The lower concentration samples were for the Lu-doped system as indicated.

Fig. 4. Phase diagram for $(La_{1-x}Gd_x)Ru_2$, indicating the T_c and T_m curves and their intersection. The dashed line under the T_c curve is speculative; it has not been measured.